Elemental Analysis of Lead Ammunition from Arbuckle's Fort (46Gb13), Greenbrier County, West Virginia

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Elemental Analysis of Lead Ammunition from Arbuckle's Fort (46Gb13),

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I. Introduction

This monograph documents a portable X-Ray fluorescence (pXRF) study of lead ammunition from Arbuckle's Fort site (46Gb13). This study builds on the recent research by the author and others on elemental analysis using pXRF on eighteenth and early-nineteenth century military sites in the eastern United States (Seibert et al. 2014; Elliott 2016; Elliott and Seibert 2017).

Arbuckle's Fort was a militia fort located in present-day Greenbrier County, West Virginia (McBride and McBride 2014). The fort was built on property of John Keeny on Mill Creek in the Muddy Creek watershed, by militia men commanded by Captain Matthew Arbuckle. The fort was used from 1774 during Dunmore's War through 1782 in the American Revolution. The archaeological remains of Arbuckle's Fort have been extensively explored.

Small arms ammunition in America, throughout the eighteenth and early nineteenth centuries, consisted of round soft-metal balls. These were mostly lead, although archeologists have documented other metals as additives. Available small arms and related ammunition varied by military unit, and included pistols, rifles, trade guns, carbines, fowlers, and large caliber wall guns, as well as American, French and English muskets. Macroscopic identification of associated bullets alone limits battlefield interpretations. Traditional analysis documents diameter, weight, firing condition (impact evidence, rifling, worming, ramrod impact, casting evidence), alterations (chewing, cutting, carving), other post-depositional damage (rodent gnawing), and archaeological context.

Elemental analysis of lead ammunition provides another layer of research data that promises to have value for distinguishing otherwise similar appearing objects. Selected elements in the Arbuckle's Fort assemblage were examined in detail. These include Antimony, Tin, Silver, Cadmium, Copper, Nickel, and Zinc. Ratios for these elements form clusters, which may have cultural significance or may be useful in future sourcing studies.

II. Methods

Previous study of eighteenth and early- nineteenth century lead artifacts from archaeological sites provide a backdrop for the present study (Sivilich 1996, 2004, 2014, 2016; Branstner 2008). These studies explored various physical aspects and characteristics of round ball ammunition.

Portable X-Ray Fluorescence (pXRF) has been used for several decades as a non-destructive method of analyzing archaeological artifacts and sediments. A recent study by Siebert and colleagues from National Park Service, Southeast Archeological Center and Bruce Kaiser examined lead shot from Palo Alto battlefield, Mexican-American War, 1846 (Seibert et al. 2016). Their study analyzed 700 lead shot. They were able to distinguish between shot from Mexican (British Brown Bess, Indian Pattern) weapons and shot from American (Springfield Arsenal, Model 1816/1822 and 1835 muskets). The simplified result is that Mexican shot contained more silver (Ag).

In his recent book on musket balls, Daniel Sivilich (2016) presented some information on pXRF results from six musket balls from Valley Forge, Pennsylvania and 104 musket balls from Monmouth Battlefield in New Jersey. He compared the frequencies of lead, iron and tin in these balls.

On December 4 and 5, 2015 a meeting of the National Park Service and the LAMAR Institute archaeologists was held at NPS Southeastern Archeological Center in Tallahassee, Florida. This pilot study used pXRF technology to identify and characterize round ball ammunition from early sites (primarily Revolutionary War period) in the eastern states. On the advice of Bruce Kaiser, inventor of the Bruker Tracer handheld device, the archeologists attempted to gather data systematically. Data files for the study were collected with Bruker Tracer III devices. Data was collected for 180 seconds for each sample using 45 kV voltage and 20 µA and Bruker's Green filter (Ti/Al). No vacuum was employed. This study demonstrated that Portable X-ray Florescence (pXRF) is a useful technology in distinguishing round ball assemblages from eighteenth and early nineteenth century sites in the eastern United States. This pilot study gathered elemental data on 440 round metal balls through a systematic data collection protocol. This sample was obtained from 14 different archeological sites from the U.S. Eastern seaboard with emphasis on the southeast. The sample spans the early eighteenth through early nineteenth centuries and it covers Native American and Euro-American towns, as well as French and Indian War, Revolutionary War, Indian Wars, and War of 1812 sites.

These data demonstrated that Antimony (Sb) and Tin (Sn) are very important elements for measuring differences in round balls. These two elements are common components of pewter.

The preliminary findings from the pilot study demonstrated that Portable X-ray Florescence (pXRF) can be a useful technology in distinguishing round ball assemblages from eighteenth and early nineteenth century sites in the eastern United States (Elliott 2016). Bruce Kaiser confounded the group by announcing a new and improved filter for the Bruker Tracer, which he

called the “Black Filter”. This filter had the addition of a thin copper sheet and was designed to reduce the masking effect caused by lead in the round balls. Using the new Black Filter, the group then proceeded to sample 72 lead balls from a variety of sites. The goal of the group was to solidify the pXRF data collection protocol so that an international database can be created and maintained. The group agreed that the database should be housed and maintained by the National Park Service. We also agreed that the breadth of the database should be widened to include the international community.

A follow-up *Get the Lead Out* workshop was held in Savannah, Georgia in 2017. Researchers were invited to bring their lead samples and pXRF datasets for analysis. The results of the workshop were summarized by Elliott and Seibert (2017). While a number of lead balls have been included, the lead study is lacking elemental data on eighteenth and early nineteenth century lead sources. A pXRF study of those lead sources would further strengthen the value of this database in understanding those relatively anonymous round bullets that are the building blocks of conflict studies. Collecting lead samples from early mines in both America, Great Britain and Europe is a high priority task.

Early lead mines have been documented at numerous locations in North America. These include Connecticut (Marteka 2009), Kentucky (Filson 2017), Massachusetts (Nash 1827), New Jersey (New Jersey Geological Society 1893), New York (Sims and Hotz 1951), Pennsylvania (Roberdeau 1778; Columbian Magazine 1788; Weatherill et al. 1826; Mitchell 1855; Stapleton 1971; FortRoberdeau.org 2014), Virginia (McGavock Papers 1760-1888; Currier 1935; Austin 1977; Whisonant 1996; Foley and Craig 1989), West Virginia (Kenny 1945), the Upper Mississippi Valley (Austin 1804; Chandler 1829; Seeger 2008; Thwait 1895; Farquhar et al. 1995; Missouri Department of Natural Resources 2017), and other locations in North America (Board of War 1777; Continental Congress 1778; Bristed 1818; Ingalls 1907, 1908).

Archeologists can improve the lead ball information by incorporating pXRF analysis of the lead balls into existing analytical framework. The ultimate goal is to elevate the diagnostic value of round ball ammunition so that we can determine where the lead came from, who was firing the bullets, and how access to lead varied over the course of history. This now appears to be an achievable goal (Elliott and Seibert 2017). Researchers are encouraged to provide input in improving this database.

Archeologists have made significant advances in musket ball analysis and interpretation over the past several decades. Musket ball diameters, represented in calibers (hundredths of inches) generally are associated with the following arms:

- American Long Rifle- .38-.51
- Fusil, American Musket, Long Rifle, Fowling Gun- .52-.59
- French Standard- .60-.66
- British Standard- .67-.74

Buck shot ranging between .29-.35 caliber were used by the Americans in buck-and-ball loads in smoothbore muskets. These were prepared paper cartridge loads that contained one large ball and two to three buck shot. The scatter of buck shot on the battlefield provides supporting

information on the American firing patterns. Some Loyalist units also used buck-and-ball loads, so its presence is not an absolute indication of Patriot's firing. Buck shot also was used in non-military contexts for hunting.

Specific Methods Employed in the Elemental Analysis

Elemental data collection for the Arbuckle's Fort sample was conducted by Daniel T. Elliott at his laboratory in Rincon, Georgia. Samples were collected using a Bruker III-V handheld device without vacuum for 180 seconds each using the Black filter (Ti/Al/Cu). Energy settings were 48 kV voltage, and 29 μA of current.

The initial analysis generated spectra and summary reports of the elemental composition for each sample. Selected elements were compared using ratios where the photon values for the selected element was divided by the photon values for Rhodium (Rh), which is present in the Bruker hardware. Creation of these ratios normalizes the dataset so that they can be quantitatively compared. Cluster analysis was then performed on the data set for selected elemental ratios.

III. Arbuckle's Fort Sample

It is against the previously described scientific backdrop in the *Get the Lead Out* study (Elliott and Seibert 2017) that an elemental analysis of the lead ammunition and related items from the Arbuckle's Fort site was set. Ten samples from Arbuckle's Fort were examined. Appendix 1 contains spectra for each sample, a report of photons for selected elements, as well as a summary table of the photon values for potentially significant elements. The results from each sample and comparative analysis of the samples are discussed in this chapter.

Sample 46Gb13-1

Sample 46Gb13-1 is a fired lead ball from Unit 49, Zone 1. It weighed 12.6 g. This sample yielded more than 100 photons each of Cadmium, Copper, Iron, Antimony and Tin. It yielded the greatest number of photons of Zinc (N=53) of the 10 samples.

Sample 46Gb13-2

Sample 46Gb13-2 is a small lead ball from Unit 55, Feature 41. It was 0.31 in in diameter and weighed 2.6 g. This sample yielded more than 100 photons each of Copper, Iron, Antimony and Tin.

Sample 46Gb13-3

Sample 46Gb13-3 is a lead ball from Unit 59, Zone 1. It measured 0.508 inches in diameter and weighed 12.47 g. This sample yielded more than 100 photons each of Cadmium, Copper, Iron, Antimony and Tin.

Sample 46Gb13-4

Sample 46Gb13-4 is a fired lead ball from Unit 59, Zone 1. It weighed 10.01 g. This sample yielded more than 100 photons each of Cadmium, Copper, Iron, Nickel, and Tin. It yielded the greatest number of photons of Nickel (N=150) of the 10 samples.

Sample 46Gb13-5

Sample 46Gb13-5 is a spent lead ball from Unit 68, Zone 1. It weighed 9.74 g. This sample yielded more than 100 photons each of Cadmium, Copper, Iron, Nickel, Antimony and Tin. It yielded the greatest number of photons of Iron (N=543), Antimony (N=7,803) and Tin (N=12,156) of the 10 samples.

Sample 46Gb13-6

Sample 46Gb13-6 is a lead ball from Unit 84, Zone 1. It measured 0.351 inches in diameter and weighed 3.8 g. This sample yielded more than 100 photons each of Cadmium, Copper, Iron, Antimony and Tin.

Sample 46Gb13-7

Sample 46Gb13-7 is a fired lead ball from Unit 93, Zone 1. It weighed 9 g. It yielded more than 100 photons each of Silver, Cadmium, Copper, Iron, Nickel, and Tin. It yielded the greatest number of photons of Cadmium (N=245) and Copper (N=370) of the 10 samples.

Sample 46Gb13-8

Sample 46Gb13-8 is a lead ball from Unit 93, Zone 1. It measured 0.477 inches in diameter and weighed 10.2 g. It yielded more than 100 photons each of Cadmium, Copper, Iron and Tin.

Sample 46Gb13-9

Sample 46Gb13-9 is a lead ball from Unit 96, Zone 1. It measured 0.409 inches in diameter and weighed 6.6 g. It yielded more than 100 photons each of Silver, Cadmium, Copper, Iron, Nickel, Antimony and Tin. It yielded the greatest number of photons of Silver (N=132) of the 10 samples.

Sample 46Gb13-10

Sample 46Gb13-10 is a small, fired lead ball from Unit 99, Zone 1. It weighed 3.2 g. It yielded more than 100 photons each of Silver, Cadmium, Copper, Iron, Nickel and Tin.

Antimony

The current dataset from the Arbuckle's Fort site contains information on several elements that are now recognized as important elements in the differentiation of the elemental characterization of round ball ammunition. Each of these elements is discussed.

Antimony (Sb) is a silvery white, brittle metalloid with the atomic number 51 (Butterman and Carlin 2004; Royal Society of Chemistry 2017). It occurs with lead ores. Antimony has a high melting point (1170°F) compared to lead. It has a value of 3 on Mohs hardness scale. In early America, Antimony was a key minor ingredient in the alloy pewter. It served to harden and strengthen the pewter.

Antimony photon- (SbK12) values were examined. Antimony was a significant component of the lead ball samples. Antimony values ranged from a low of 30 photons in Sample 46Gb13-7 to a high of 7803 in Sample 46Gb13-5. Five of the 10 samples had values greater than 100 photons.

The Arbuckle's Fort Rhodium ratio data for Silver, Antimony and Tin then was compared to the other 932 samples that were gathered previously for the lead ball study. Sample 46Gb13-5 was the third highest ranking in Antimony (Sb)/Rhodium (Rh) ratio in the entire lead ball study. Other high ranking samples included 46Gb13-9 and 46Gb13-3, which ranked 64th and 68th, respectively. Of the Arbuckle's Fort samples, 46Gb13-7 ranked the lowest as the 562nd of the 942 samples.

Cadmium

Cadmium (Cd) is a soft, ductile metal with the atomic number 48 (Butterman and Plachy 2004; International Cadmium Association 2017). Cadmium occurs as an impurity in lead ores. Cadmium has a melting point of 610°F, which is slightly lower than that of lead. It has a value of 2 on Mohs hardness scale.

Cadmium was a significant component of the Arbuckle Fort lead ball samples. Cadmium photon (CdK12) values were examined. These ranged from a low of 69 photons in Sample 46Gb13-2 to a high of 245 in Sample 46Gb13-7. Nine of the 10 samples had values greater than 100 photons.

Sample 46Gb13-8 had the highest Cadmium (Cd)/Rhodium (Rh) ratio in the entire lead ball study (N=942), ranking 27th with a ratio of 7.5. Sample 46Gb13-6 had the lowest ranking, with a ratio of 2.646. -It was ranked 190th.

Copper

Copper (Cu) is a malleable reddish-gold metal with the atomic number 29 (Doebrich 2009:1-4). It often occurs with lead ores. Copper has a very high melting point (1984°F) compared to lead. It has a value of 3 on Mohs hardness scale.

Copper was a significant component of the lead ball sample. Copper photon (CuK12) values were examined. These ranged from a low of 155 photons in Sample 46Gb13-2 to a high of 370 in Sample 46Gb13-7. All 10 samples had values greater than 100 photons.

Sample 46Gb13-8 had the highest Copper (Cu)/Rhodium (Rh) ratio in the entire lead ball study (N=942), ranking 39th with a ratio of 12.269. Sample 46Gb13-10 had the lowest ranking, with a ratio of 4.364, making it 254th.

Nickel

Nickel (Ni) is a silvery-white lustrous metal with the atomic number 28 (Nickel Institute 2017). Nickel has a very high melting point (2646°F) compared to lead. It has a value of 4.0 on Mohs hardness scale.

Nickel photon (NiK12) values were examined. Nickel was a moderately significant component in the lead ball samples. These ranged from a low of 75 photons in Sample 46Gb13-8 to a high of 150 in Sample 46Gb13-4. Five of 10 samples had values greater than 100 photons.

Sample 46Gb13-4 had the highest Nickel (Ni)/Rhodium (Rh) ratio in the entire lead ball study (N=942), ranking 100th with a ratio of 4.545. Sample 46Gb13-6 had the lowest ranking, with a ratio of 1.771, producing a ranking of 247th.

Silver

Silver (Ag) is a precious silver metal with the atomic number 47 (Butterman and Hilliard 2004). Silver has a high melting point (1761°F) compared to lead. It has a value of 2.5 on Mohs hardness scale. It commonly occurs with lead ores.

Silver was a minor component of the lead ball sample. Silver photon (AgK12) values were examined. These ranged from a low of 50 photons in Sample 46Gb13-6 to a high of 132 in Sample 46Gb13-9. Three of the 10 samples had values greater than 100 photons.

Sample 46Gb13-9 was the 73rd highest ranking in Silver (Ag)/Rhodium (Rh) ratio in the entire lead ball study. Sample 46Gb13-6 was the lowest ranking of the Arbuckle's Fort samples, ranking 293rd in the entire lead ball study.

Tin

Tin (Sn) is a soft, white metal with the atomic number 50 (Calvert 2002). It occurs with lead ores. Tin has a melting point of 449°F, which is lower than that of lead. It has a value of 1.5 on Mohs hardness scale. Tin is a major component of pewter alloy.

Tin was a significant component of the lead ball samples. Tin photon (SnK12) values were examined. These ranged from a low of 176 photons in Sample 46Gb13-2 to a high of 12156 in Sample 46Gb13-5. All 10 samples had values greater than 100 photons.

Sample 46Gb13-5 was the 20th highest ranking in Tin (Sn)/Rhodium (Rh) ratio in the entire lead ball study. Other high ranking samples included 46Gb13-9 and 46Gb13-3, ranking 27th and 28th respectively. Sample 46Gb13-7 was the lowest ranking of the Arbuckle's Fort samples, ranking 378th in the entire lead ball study.

Zinc

Zinc (Zn) is a lustrous metal with the atomic number 30 (International Zinc Association 2017). It is found with lead ores. Zinc has a high melting point (787°F). Zinc has a value of 2.5-3 on Mohs hardness scale.

Zinc is a minor component of the lead ball sample. Zinc photon (ZnK12) values were examined. These ranged from a low of 21 photons in Samples 46Gb13-6 and 46Gb13-10 to a high of 53 in Sample 46Gb13-1. None of the 10 samples had values greater than 100 photons.

Sample 46Gb13-1 had the highest Zinc (Zn)/Rhodium (Rh) ratio in the Arbuckle's Fort sample. in entire lead ball study (N=942), ranking 57th with a ratio of 1.893. Sample 46Gb13-6 had the lowest ranking in the Arbuckle's Fort sample, with a ratio of 0.438. It was 268th in the entire lead ball study.

Relationships between Selected Elements in the Samples

We examined the relationship between Silver (Ag), Antimony (Sb) and Tin (Sn) in the Arbuckle's Fort data. This was accomplished by expressing each as a ratio relative to the Rhodium (Rh) values, which represents a constant in the Bruker Tracer hardware. Table 1 shows a comparison of Silver (Ag), Antimony (Sb) and Tin (Sn) Photons in the Arbuckle's Fort assemblage. Samples 46Gb13-3, 46Gb13-5 and 46Gb13-9 have significant quantities of Antimony and Tin. This may indicate that pewter was melted with the lead in these examples.

Only three samples contained more than 100 photons of silver. This likely indicates that silver was not an intentional additive to the lead ball mixture but was incidental in the ore.

Figure 1 is a scatterplot of Silver/Rhodium, Antimony/Rhodium and Tin/Rhodium ratios for the Arbuckle's Fort assemblage.

Table 1. Comparison of Silver (Ag), Antimony (Sb) and Tin (Sn) Photons, Arbuckle's Fort.

Sample	Sb K12	Sn K12	Ag K12	Sb/Rh	Sn/Rh	Ag/Rh
46Gb13-1	118	1554	71	4.21429	55.5	2.5357
46Gb13-2	72	176	97	2.76923	6.76923	3.7308
46Gb13-3	487	5627	91	13.9143	160.771	2.6
46Gb13-4	50	1019	98	1.51515	30.8788	2.9697
46Gb13-5	7803	12156	64	159.245	248.082	1.3061
46Gb13-6	148	2454	50	3.08333	51.125	1.0417
46Gb13-7	30	231	102	0.55556	4.27778	1.8889
46Gb13-8	33	209	82	1.26923	8.03846	3.1538
46Gb13-9	490	5828	132	14.4118	171.412	3.8824
46Gb13-10	31	230	126	0.70455	5.22727	2.8636
Average	**926.2**	**2948.4**	**91.3**	**20.1682**	**74.2081**	**2.5973**

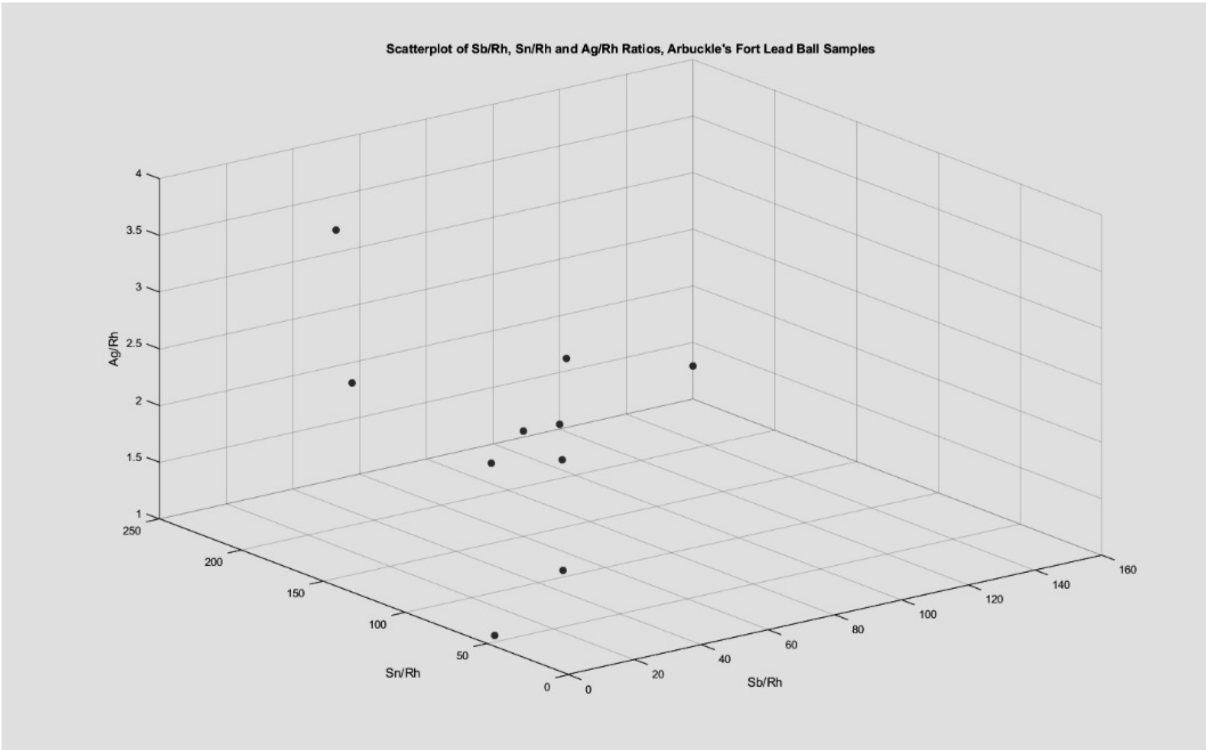

Figure 1. Scatterplot of Silver, Antimony and Tin Ratios, Arbuckle's Fort.

Cluster analysis identified five clusters (or segments) in these data for Antimony, Tin and Silver, which are detailed in Table 2 and Figure 2. Cluster 3 was prevalent, represented by four samples. It included samples 46Gb13-2, 4, 8 and 10. This cluster had mean (centroid) values of 1.56 for Sb/Rh, 12.73 for Sn/Rh, and 3.18 for Ag/Rh.

Cluster 1 was next most common, represented by samples 46Gb13-1, 3 and 7. This cluster had mean values of 6.23 for Sb/Rh, 73.52 for Sn/Rh and 2.34 for Ag/Rh.

Clusters 2, 4 and 5 were each represented by a single sample. These were samples 46Gb13-5, 9 and 6, respectively. Cluster 2 had mean values of 159.24 for Sb/Rh, 248.08 for Sn/Rh and 1.31 for Ag/Rh. Cluster 4 had mean values of 14.41 for Sb/Rh, 171.41 for Sn/Rh and 3.88 for Ag/Rh. Cluster 5 had mean values of 3.08 for Sb/Rh, 51.13 for Sn/Rh and 1.04 for Ag/Rh.

Table 2. Output for Five Clusters, Sb/Rh, Sn/Rh and Ag/Rh, Arbuckle's Fort Samples.

Output for FIVE Clusters/Segments							
Mean/Centroid	***Sb/Rh***	***Sn/Rh***	***Ag/Rh***	***0***	***0***	***0***	***0***
Segment 1	6.23	73.52	2.34				
Segment 2	159.24	248.08	1.31				
Segment 3	1.56	12.73	3.18				
Segment 4	14.41	171.41	3.88				
Segment 5	3.08	51.13	1.04				
AVERAGE	20.17	74.21	2.60				
Respondents	**Number**	**%**	**SSE/Segment**				
Segment 1	3	30.0%	12827.6				
Segment 2	1	10.0%	0.0	**SSE Total**	**3.2**		
Segment 3	4	40.0%	0.0				
Segment 4	1	10.0%	0.0				
Segment 5	1	10.0%	0.0				
TOTAL	10	100.0%					

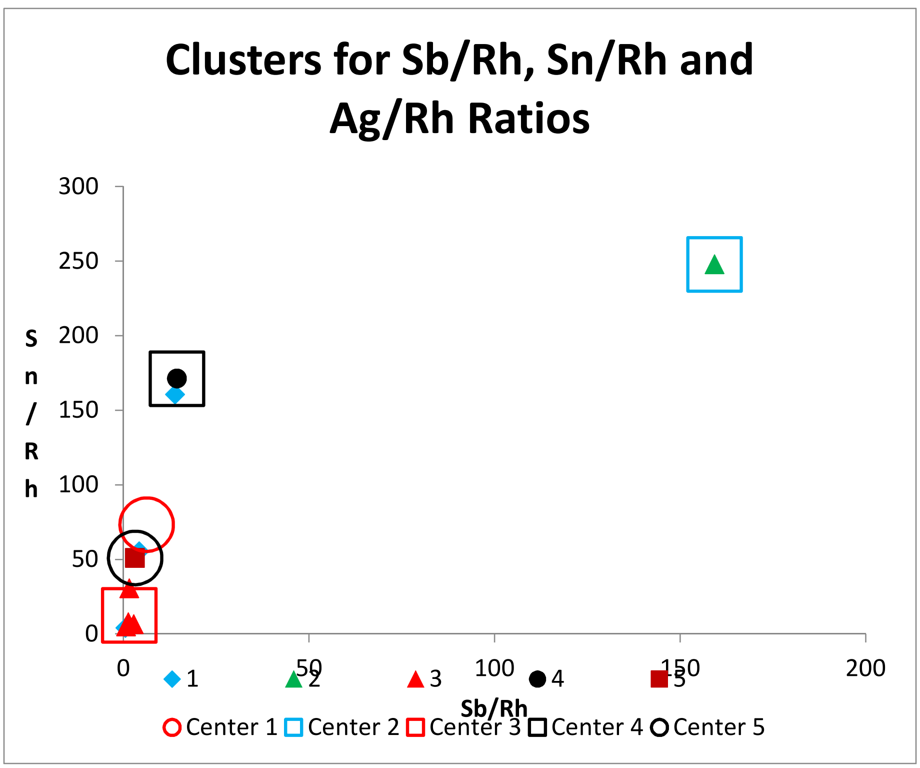

Figure 2. Central Means Chart for Five Clusters, Sb/Rh, Sn/Rh and Ag/Rh Ratios, Arbuckle's Fort Samples.

Cluster analysis identified five clusters in these data for Cadmium, Copper, Nickel and Zinc, which are detailed in Table 3 and Figure 3. Cluster 1 was most common, represented by samples 46Gb13-5, 6,7 and 10. This cluster had mean values of 3.33 for Cd/Rh, 5.69 for Cu/Rh, 2.31 for Ni/Rh and 0.53 for Zn/Rh.

Cluster 4 was next most common represented by samples 46Gb13-1. 3 and 9. This cluster had mean values of 4.55 for Cd/Rh, 9.29 for Cu/Rh, 3.02 for Ni/Rh and 1.57 for Zn/Rh.

Clusters 2, 3 and 5 were each represented by a single sample. These were samples 46Gb13-8, 4 and 2, respectively. Cluster 2 had mean values of 7.5 for Cd/Rh, 12.27 for Cu/Rh, 2.88 for Ni/Rh and 1.81 for Zn/Rh. Cluster 3 had mean values of 5.79 for Cd/Rh, 9.91 for Cu/Rh, 4.55 for Ni/Rh and 0.94 for Zn/Rh. Cluster 5 had mean values of 2.65 for Cd/Rh, 5.96 for Cu/Rh, 3.15 for Ni/Rh and 1.11 for Zn/Rh.

Table 3. Output for Five Clusters, Cd/Rh, Cu/Rh, Ni/Rh and Zn/Rh, Arbuckle's Fort Samples.

Output for FIVE Clusters/Segments							
Mean/Centroid	***Cd/Rh***	***Cu/Rh***	***Ni/Rh***	***Zn/Rh***	***0***	***0***	***0***
Segment 1	3.33	5.69	2.31	0.53			
Segment 2	7.50	12.27	2.88	1.81			
Segment 3	5.79	9.91	4.55	0.94			
Segment 4	4.55	9.29	3.02	1.57			
Segment 5	2.65	5.96	3.15	1.54			
AVERAGE	4.29	7.88	2.89	1.11			
Respondents	**Number**	**%**	**SSE/Segment**				
Segment 1	4	40.0%	5.7				
Segment 2	1	10.0%	0.0	**SSE Total**	**6.2**		
Segment 3	1	10.0%	0.0				
Segment 4	3	30.0%	2.3				
Segment 5	1	10.0%	0.0				
TOTAL	10	100.0%					

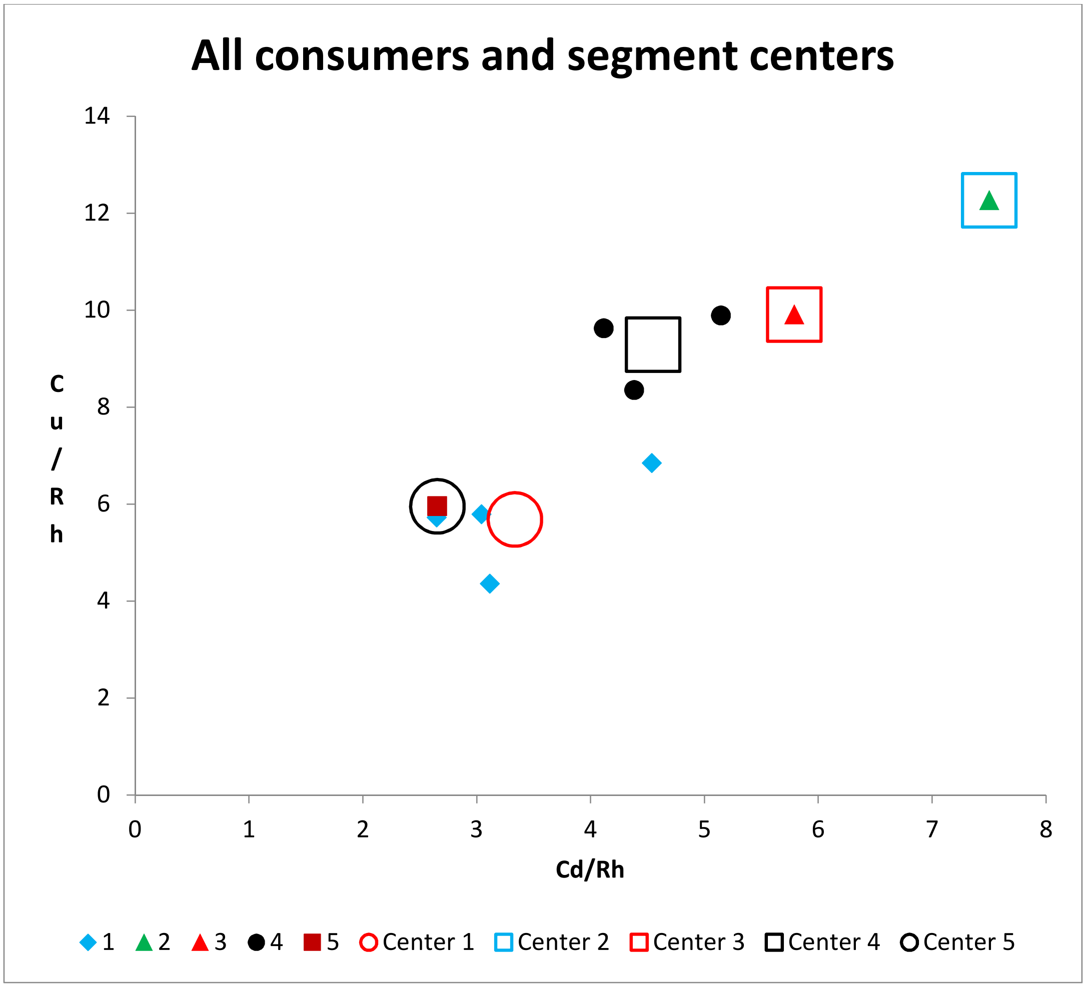

Figure 3. Central Means Chart for Five Clusters, Cd/Rh, Cu/Rh. Ni/Rh, and Zn/Rh Ratios, Arbuckle's Fort Samples.

IV. Summary

Ten pieces of lead ammunition from the Arbuckle's Fort archaeological site (46Gb13) in Greenbrier County, West Virginia were analyzed for their elemental composition using Portable X-ray Fluorescence (pXRF) technology. Arbuckle's Fort is a Patriot militia military fortification that existed from about 1774 to 1783. These artifacts were professionally excavated and have a secure cultural context dating to that period of the late eighteenth century (McBride and McBride 2014).

While the sample set was small, which limited statistical manipulations, some comparisons in the data were offered. The samples contain impurities of elements other than lead, which are consistent with previously studied collections from Patriot sites in eastern North America (Elliott and Seibert 2017). The significant incidence of Antimony (Sb) and Tin (Sn), both of which are components of pewter suggests that pewter was combined with the lead during bullet manufacture. The Arbuckle's Fort lead ball dataset is a significant addition to the growing database for the study of early lead ammunition.

The significant presence of other elements, including Cadmium (Cd), Copper (Cu), Nickel (Ni), Silver (Ag), and Zinc (Zn) were observed. The cultural significance of the presence of these other elements, if any, remains unclear at present. All of these elements are found naturally in association with lead ore deposits.

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Appendix 1.

Elemental Analysis Data, Arbuckle's Fort

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:54:26 PM

Serial number:
Spectrum: 46gb13-1@120318_092540
Method: Lead balls (Bayes)
Count rate: 1668 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:23:04 PM
Live time: 166 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	32	90
Mn	K12	0.00	34	109
Fe	K12	0.00	435	119
Co	K12	0.00	57	129
Ni	K12	0.00	84	142
Cu	K12	0.00	277	173
Zn	K12	0.00	53	192
Ga	K12	0.00	679	284
As	K12	0.00	2698	623
Sr	K12	0.00	280	270
Y	K12	0.00	1908	362
Zr	K12	0.00	379	209
Nb	K12	0.00	2	19
Rh	K12	0.00	28	50
Rh	L1	0.00		242
Pd	K12	0.00	70	74
Pd	L1	0.00	32	219
Ag	K12	0.00	71	113
Ag	L1	0.00	1	209
Cd	K12	0.00	144	174
Cd	L1	0.00		183
Sn	K12	0.00	1523	333
Sn	L1	0.00	59	162
Sb	K12	0.00	118	469
Sb	L1	0.00	40	170
Pb	L1	0.00	88380	570
Pb	M1	0.00	519	293

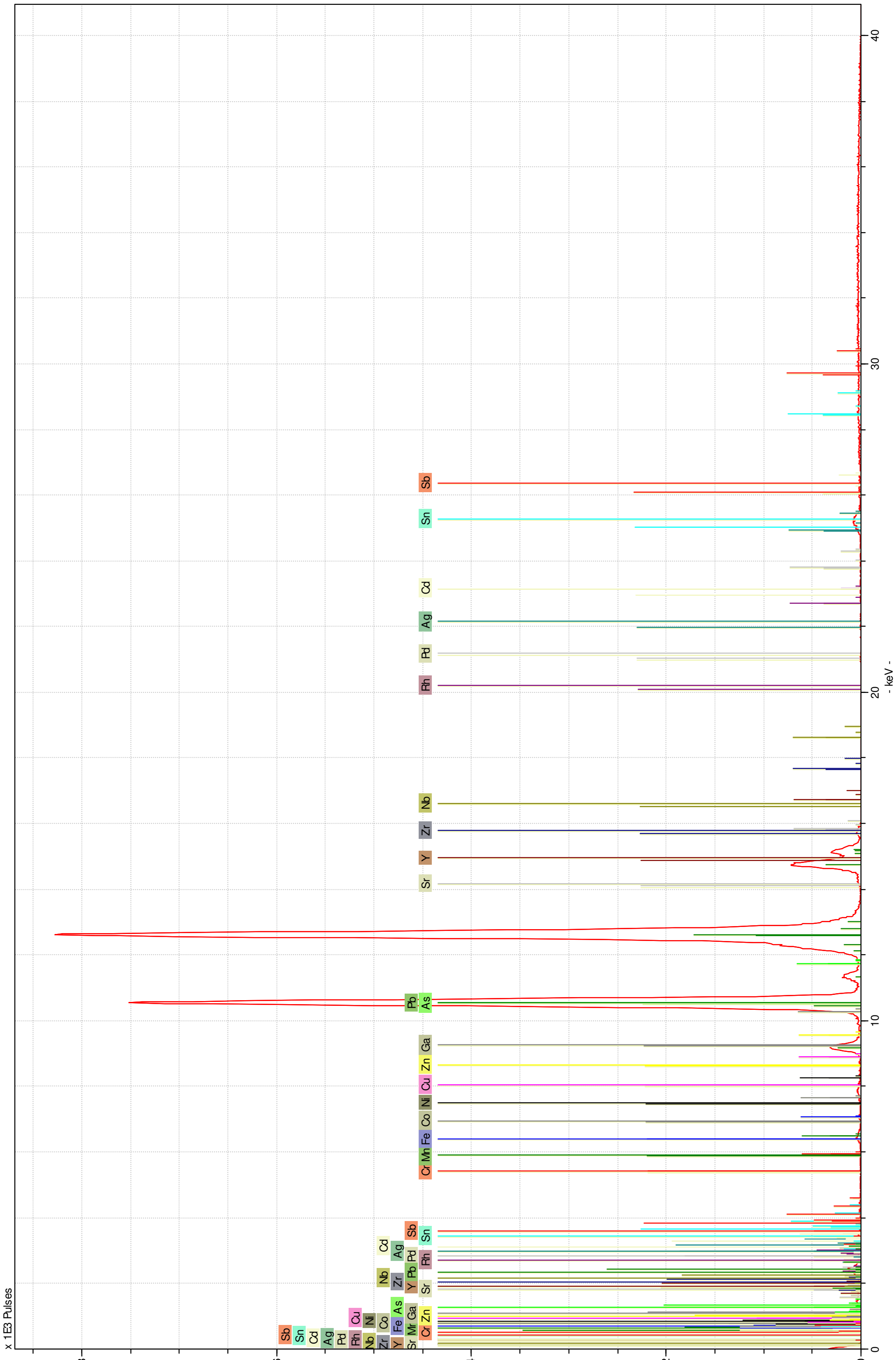

Figure 1. Spectra of Sample 46Gb13-1

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:55:25 PM

Serial number:
Spectrum: 46gb13-2@120318_092540
Method: Lead balls (Bayes)
Count rate: 729 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:27:05 PM
Live time: 174 s
Dead time: 0.0 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	11	51
Mn	K12	0.00	21	58
Fe	K12	0.00	308	62
Co	K12	0.00	19	69
Ni	K12	0.00	82	77
Cu	K12	0.00	155	92
Zn	K12	0.00	40	97
Ga	K12	0.00	253	136
As	K12	0.00	987	286
Sr	K12	0.00	171	96
Y	K12	0.00	806	144
Zr	K12	0.00	124	90
Nb	K12	0.00	5	15
Rh	K12	0.00	26	34
Rh	L1	0.00		195
Pd	K12	0.00	30	42
Pd	L1	0.00	10	180
Ag	K12	0.00	97	67
Ag	L1	0.00	11	178
Cd	K12	0.00	69	87
Cd	L1	0.00		162
Sn	K12	0.00	176	226
Sn	L1	0.00	14	136
Sb	K12	0.00	72	351
Sb	L1	0.00	31	137
Pb	L1	0.00	36246	262
Pb	M1	0.00	85	231

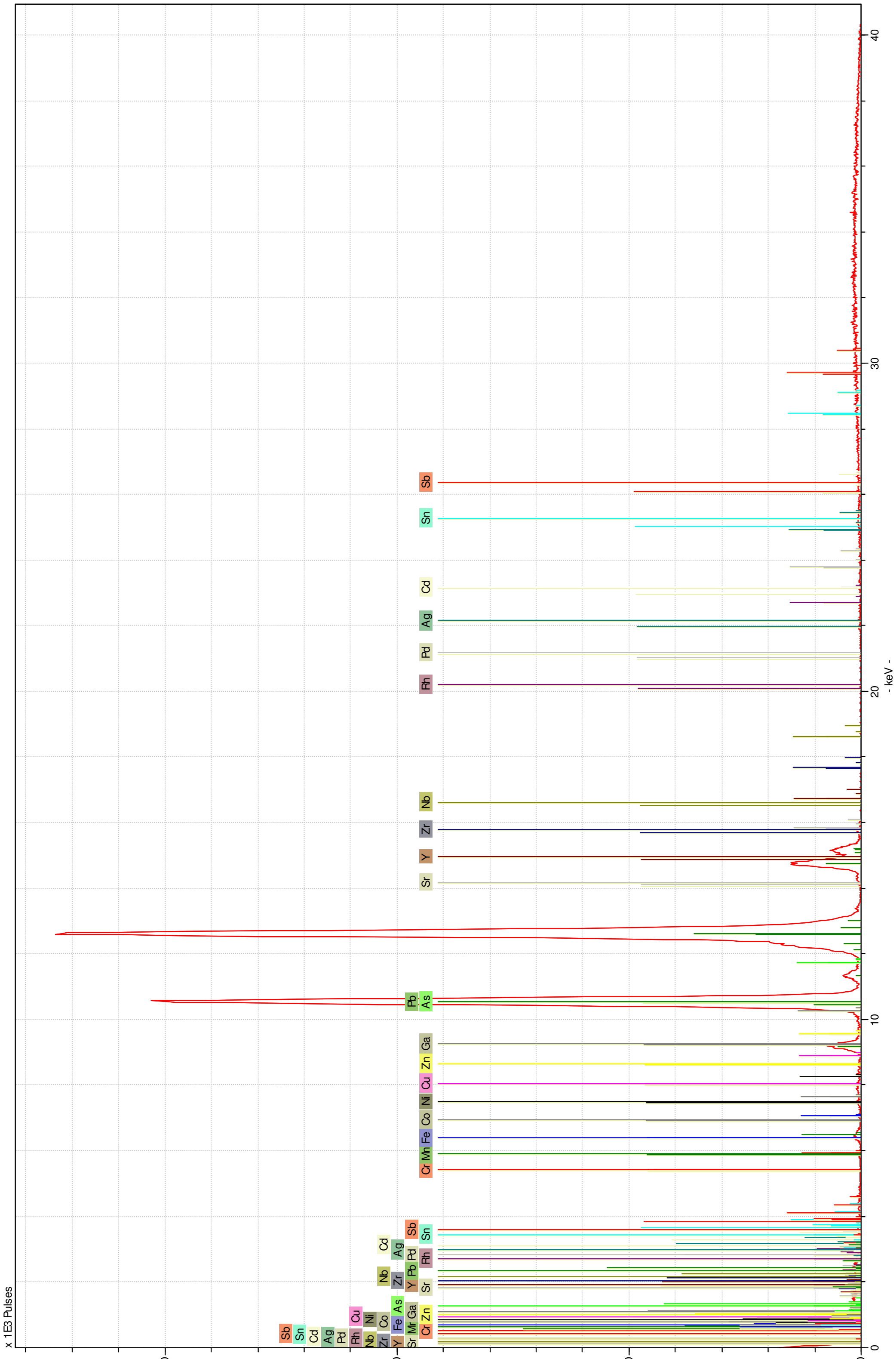

Figure 2. Spectra of Sample 46Gb13-2.

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:56:51 PM

Serial number:
Spectrum: 46gb13-3@120318_092540
Method: Lead balls (Bayes)
Count rate: 1656 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:31:39 PM
Live time: 166 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	37	93
Mn	K12	0.00	35	91
Fe	K12	0.00	292	115
Co	K12	0.00	33	142
Ni	K12	0.00	93	155
Cu	K12	0.00	337	162
Zn	K12	0.00	49	147
Ga	K12	0.00	611	261
As	K12	0.00	2724	616
Sr	K12	0.00	253	298
Y	K12	0.00	1942	368
Zr	K12	0.00	383	203
Nb	K12	0.00	9	27
Rh	K12	0.00	35	52
Rh	L1	0.00		266
Pd	K12	0.00	70	65
Pd	L1	0.00	20	245
Ag	K12	0.00	91	115
Ag	L1	0.00		243
Cd	K12	0.00	144	187
Cd	L1	0.00		229
Sn	K12	0.00	5627	363
Sn	L1	0.00	35	196
Sb	K12	0.00	487	538
Sb	L1	0.00	53	193
Pb	L1	0.00	86168	563
Pb	M1	0.00	563	318

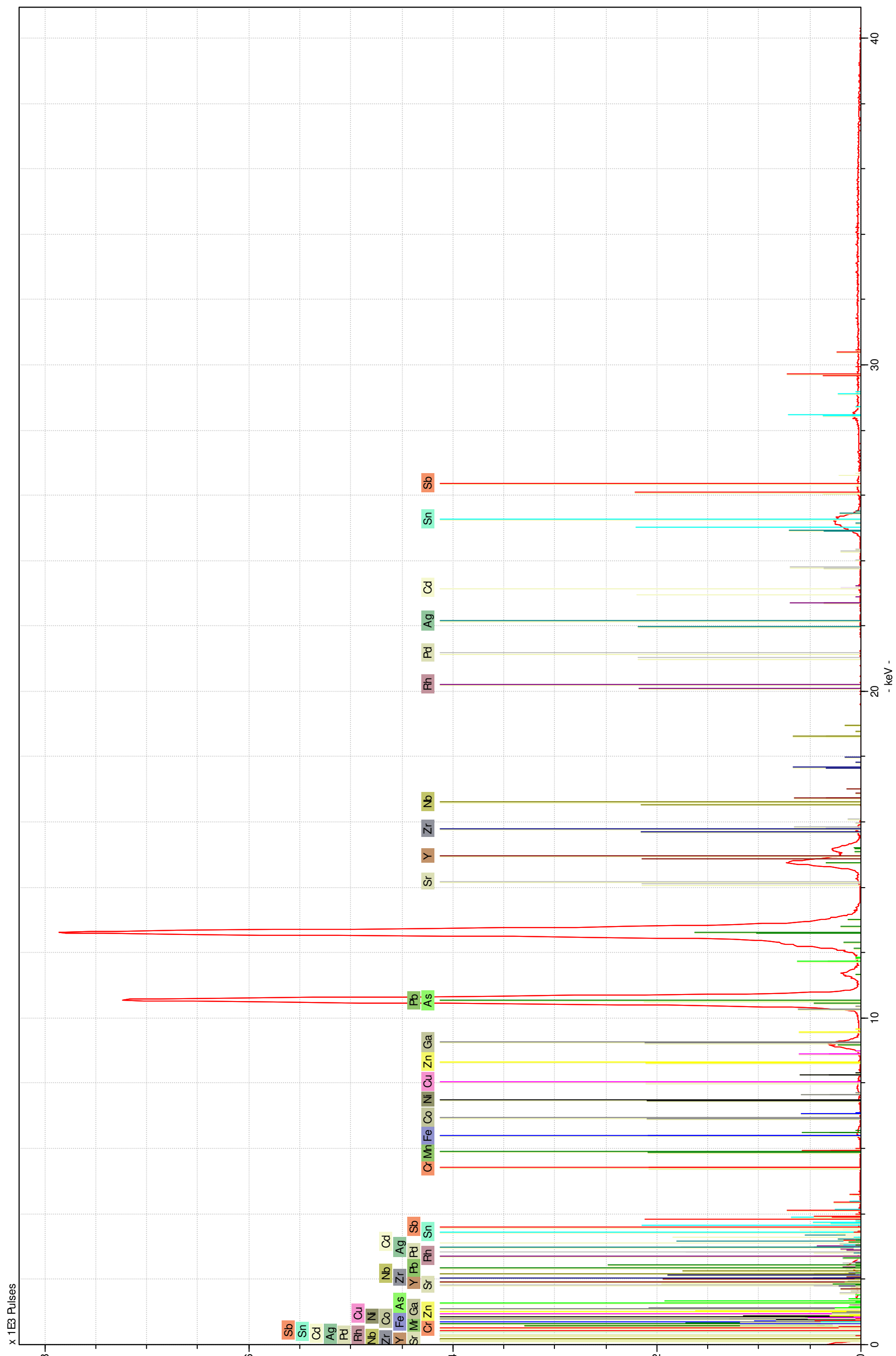

Figure 3. Spectra of Sample 46Gb13-3.

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:57:05 PM

Serial number:
Spectrum: 46gb13-4@120318_092540
Method: Lead balls (Bayes)
Count rate: 1849 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:37:11 PM
Live time: 164 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	36	108
Mn	K12	0.00	37	98
Fe	K12	0.00	406	109
Co	K12	0.00	48	140
Ni	K12	0.00	150	164
Cu	K12	0.00	327	195
Zn	K12	0.00	31	219
Ga	K12	0.00	833	329
As	K12	0.00	3038	700
Sr	K12	0.00	305	333
Y	K12	0.00	2189	403
Zr	K12	0.00	394	214
Nb	K12	0.00	6	35
Rh	K12	0.00	33	48
Rh	L1	0.00	1	274
Pd	K12	0.00	93	66
Pd	L1	0.00		257
Ag	K12	0.00	98	106
Ag	L1	0.00	9	261
Cd	K12	0.00	191	148
Cd	L1	0.00	1	248
Sn	K12	0.00	1019	296
Sn	L1	0.00		218
Sb	K12	0.00	50	449
Sb	L1	0.00	24	219
Pb	L1	0.00	98320	640
Pb	M1	0.00	590	320

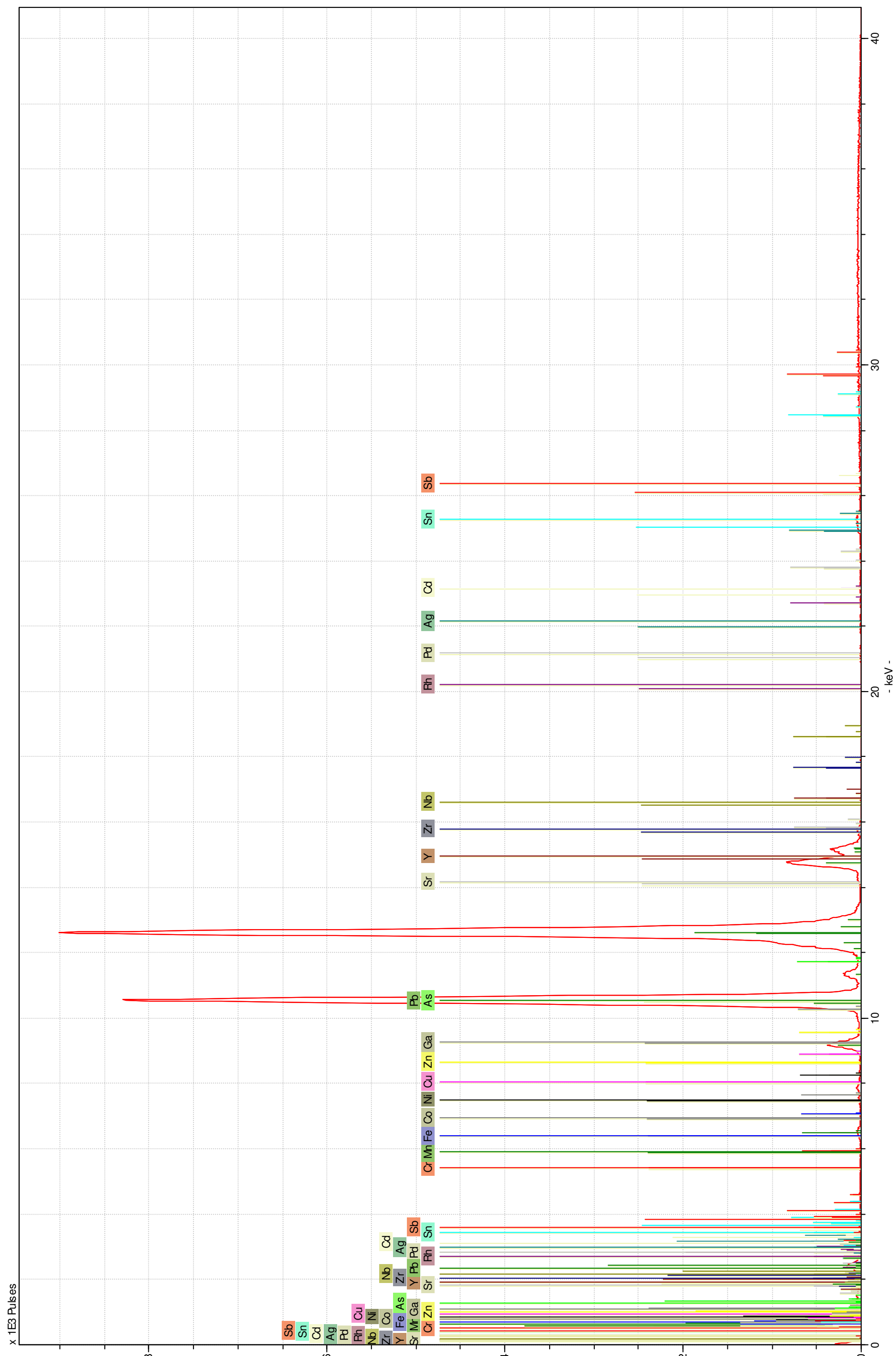

Figure 4. Spectra of Sample 46Gb13-4.

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:57:19 PM

Serial number:
Spectrum: 46gb13-5@120318_092540
Method: Lead balls (Bayes)
Count rate: 1767 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:41:26 PM
Live time: 165 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	47	101
Mn	K12	0.00	39	98
Fe	K12	0.00	543	119
Co	K12	0.00	37	143
Ni	K12	0.00	122	159
Cu	K12	0.00	284	180
Zn	K12	0.00	22	185
Ga	K12	0.00	543	287
As	K12	0.00	2219	659
Sr	K12	0.00	297	272
Y	K12	0.00	2042	365
Zr	K12	0.00	436	217
Nb	K12	0.00	10	59
Rh	K12	0.00	49	55
Rh	L1	0.00		357
Pd	K12	0.00	72	105
Pd	L1	0.00	28	333
Ag	K12	0.00	64	162
Ag	L1	0.00		332
Cd	K12	0.00	149	284
Cd	L1	0.00		314
Sn	K12	0.00	12156	609
Sn	L1	0.00	86	277
Sb	K12	0.00	7803	717
Sb	L1	0.00	155	276
Pb	L1	0.00	83680	604
Pb	M1	0.00	403	419

Figure 5. Spectra of Sample 46Gb13-5.

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:57:34 PM

Serial number:
Spectrum: 46gb13-6@120318_092540
Method: Lead balls (Bayes)
Count rate: 1433 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:45:22 PM
Live time: 168 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	27	93
Mn	K12	0.00	18	96
Fe	K12	0.00	256	101
Co	K12	0.00	18	118
Ni	K12	0.00	85	130
Cu	K12	0.00	275	148
Zn	K12	0.00	21	156
Ga	K12	0.00	508	243
As	K12	0.00	2269	559
Sr	K12	0.00	246	226
Y	K12	0.00	1630	282
Zr	K12	0.00	289	148
Nb	K12	0.00	1	26
Rh	K12	0.00	48	42
Rh	L1	0.00		224
Pd	K12	0.00	56	64
Pd	L1	0.00	8	202
Ag	K12	0.00	50	120
Ag	L1	0.00		197
Cd	K12	0.00	127	178
Cd	L1	0.00		188
Sn	K12	0.00	2454	344
Sn	L1	0.00	59	178
Sb	K12	0.00	148	458
Sb	L1	0.00	21	183
Pb	L1	0.00	75820	511
Pb	M1	0.00	507	276

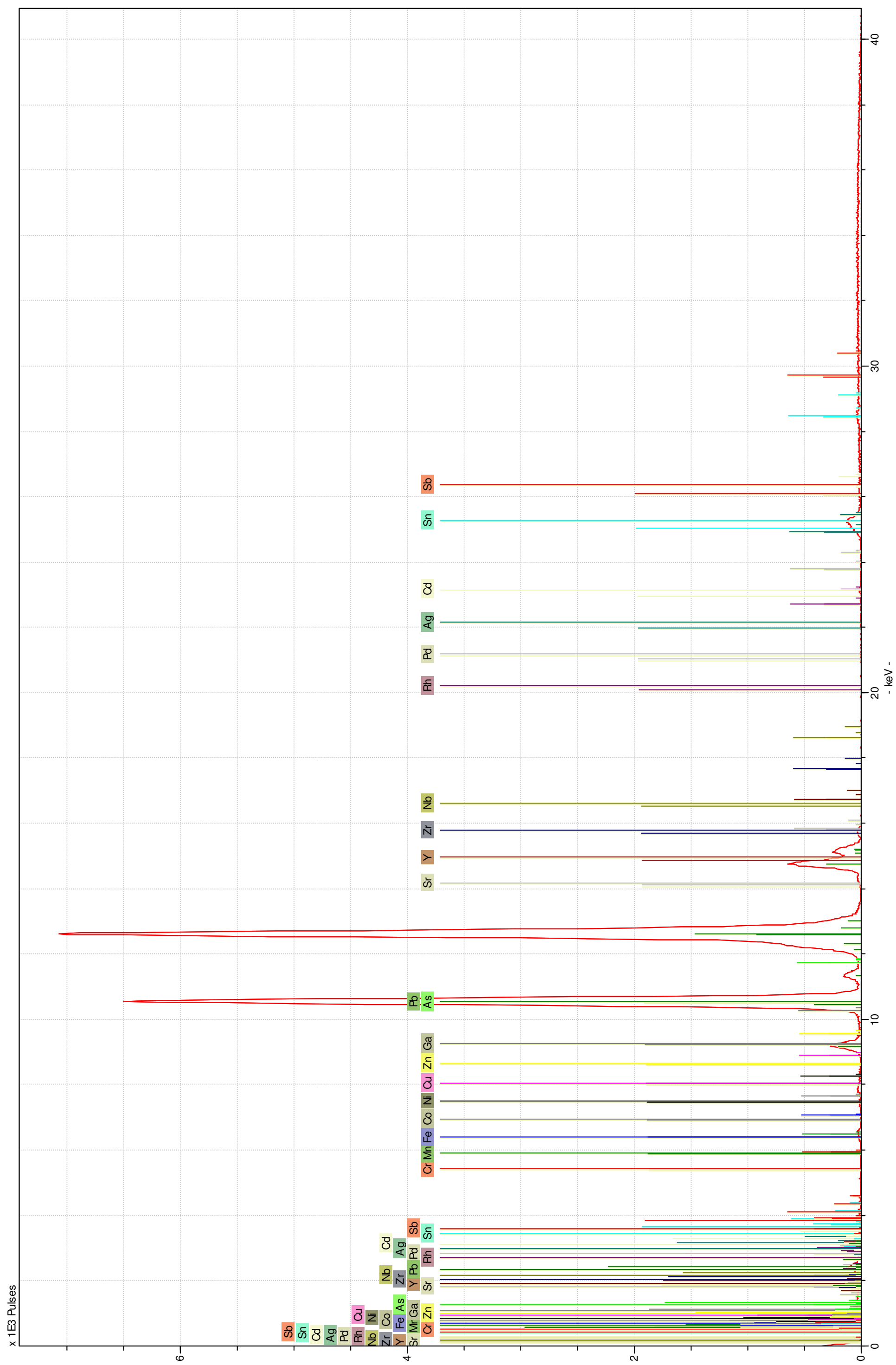

Figure 6. Spectra of Sample 46Gb13-6.

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:57:45 PM

Serial number:
Spectrum: 46gb13-7@120318_092540
Method: Lead balls (Bayes)
Count rate: 2006 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:49:23 PM
Live time: 163 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	66	116
Mn	K12	0.00	24	111
Fe	K12	0.00	355	126
Co	K12	0.00	43	144
Ni	K12	0.00	124	167
Cu	K12	0.00	370	208
Zn	K12	0.00	41	231
Ga	K12	0.00	873	369
As	K12	0.00	3463	752
Sr	K12	0.00	376	353
Y	K12	0.00	2151	445
Zr	K12	0.00	401	252
Nb	K12	0.00	5	32
Rh	K12	0.00	54	38
Rh	L1	0.00		296
Pd	K12	0.00	99	65
Pd	L1	0.00	16	268
Ag	K12	0.00	102	111
Ag	L1	0.00	1	265
Cd	K12	0.00	245	148
Cd	L1	0.00	4	253
Sn	K12	0.00	231	353
Sn	L1	0.00		230
Sb	K12	0.00	30	500
Sb	L1	0.00	19	234
Pb	L1	0.00	107278	687
Pb	M1	0.00	687	359

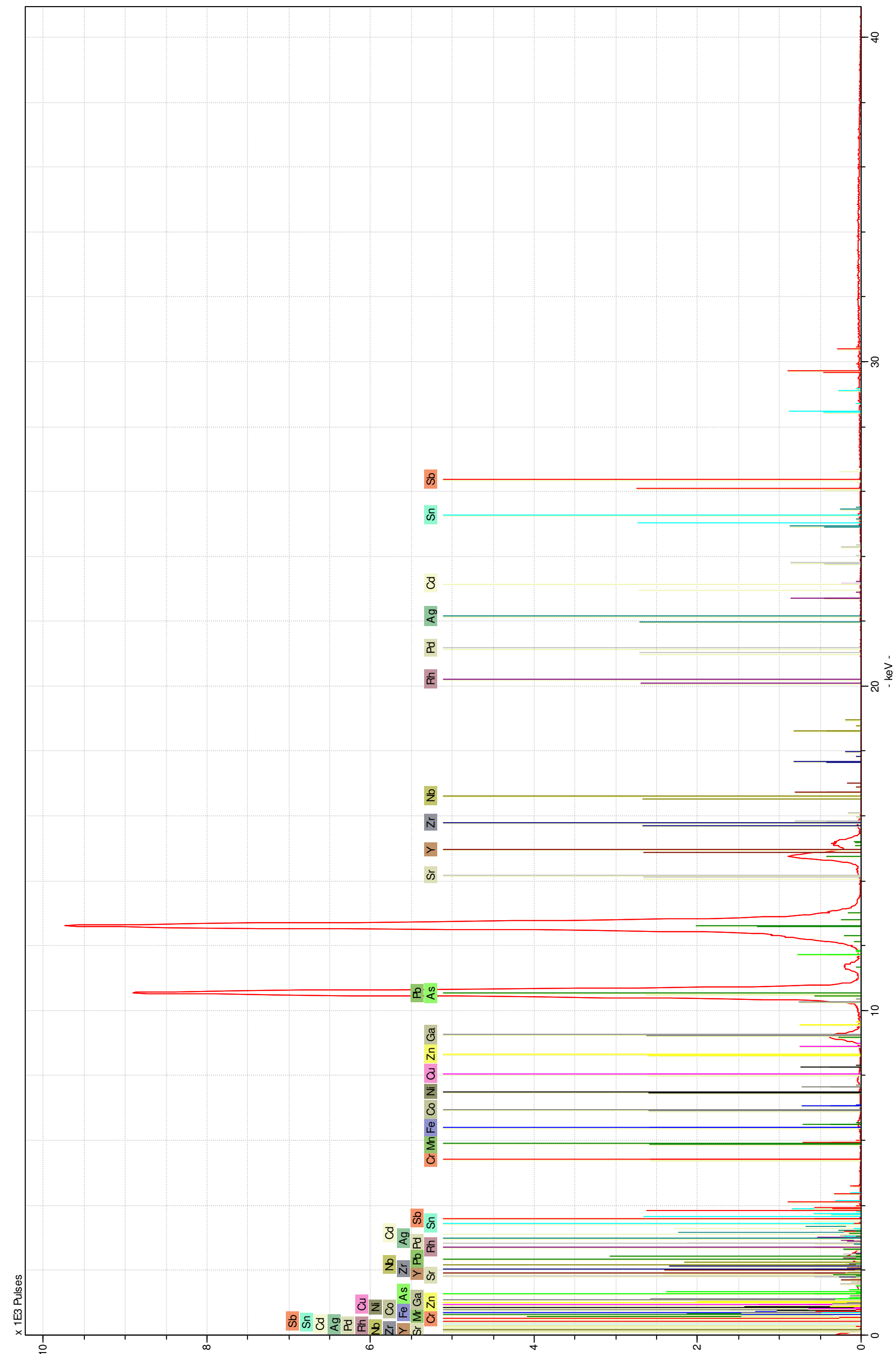

Figure 7. Spectra of Sample 46Gb13-7.

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:58:02 PM

Serial number:
Spectrum: 46gb13-8@120318_092540
Method: Lead balls (Bayes)
Count rate: 1770 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:53:23 PM
Live time: 165 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	36	114
Mn	K12	0.00	38	101
Fe	K12	0.00	316	111
Co	K12	0.00	42	146
Ni	K12	0.00	75	168
Cu	K12	0.00	319	186
Zn	K12	0.00	47	202
Ga	K12	0.00	749	302
As	K12	0.00	2716	660
Sr	K12	0.00	238	318
Y	K12	0.00	2011	386
Zr	K12	0.00	391	213
Nb	K12	0.00		29
Rh	K12	0.00	26	44
Rh	L1	0.00		248
Pd	K12	0.00	61	66
Pd	L1	0.00	10	237
Ag	K12	0.00	82	96
Ag	L1	0.00	5	244
Cd	K12	0.00	195	120
Cd	L1	0.00		235
Sn	K12	0.00	209	316
Sn	L1	0.00	11	212
Sb	K12	0.00	33	448
Sb	L1	0.00	43	215
Pb	L1	0.00	94528	605
Pb	M1	0.00	632	273

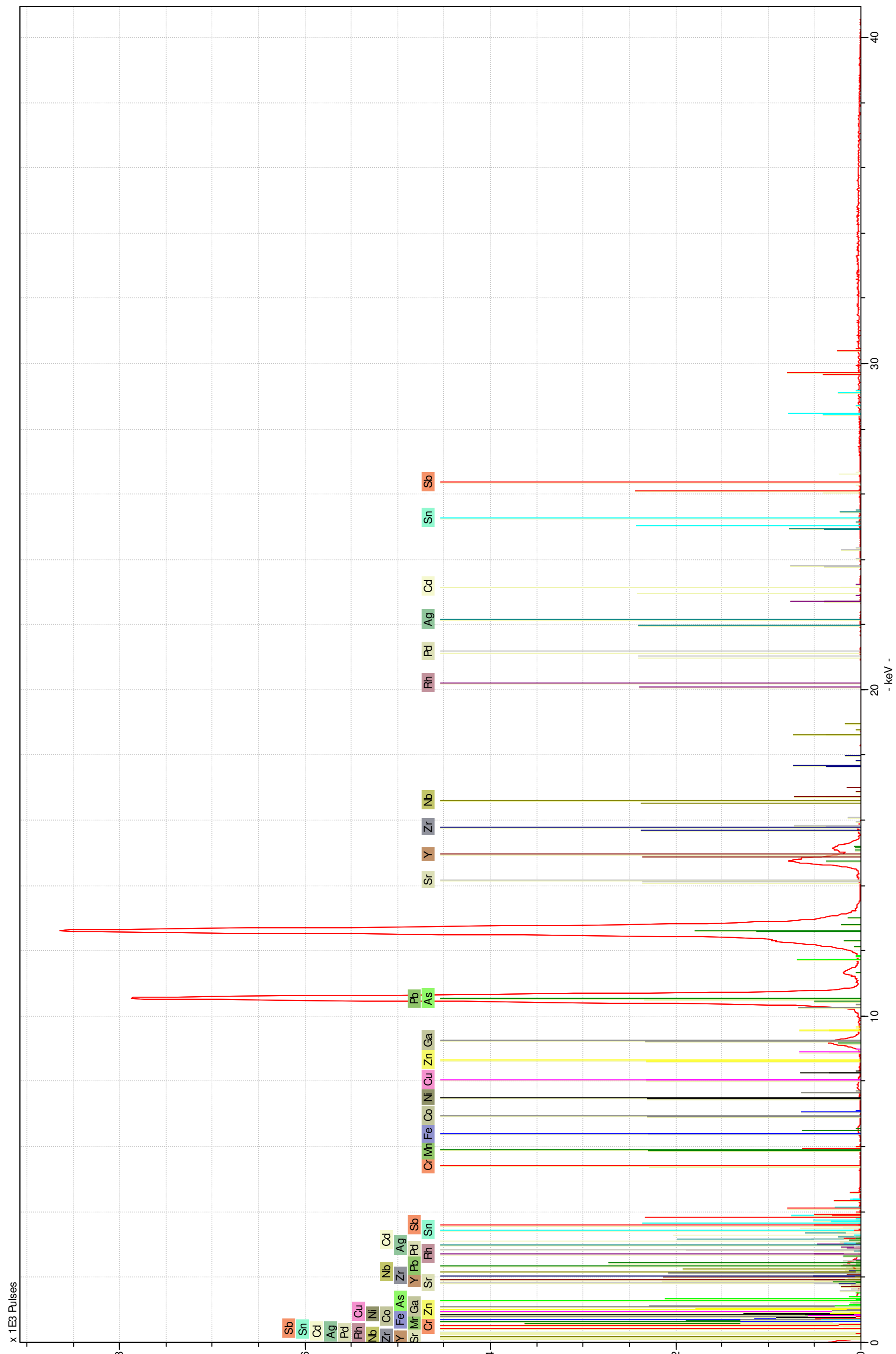

Figure 8. Spectra of Sample 46Gb13-8.

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:58:27 PM

Serial number:
Spectrum: 46gb13-9@120318_092540
Method: Lead balls (Bayes)
Count rate: 1675 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 1:57:34 PM
Live time: 166 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	79	89
Mn	K12	0.00	20	101
Fe	K12	0.00	354	107
Co	K12	0.00	12	118
Ni	K12	0.00	114	133
Cu	K12	0.00	284	156
Zn	K12	0.00	48	164
Ga	K12	0.00	696	263
As	K12	0.00	2676	609
Sr	K12	0.00	289	280
Y	K12	0.00	1925	352
Zr	K12	0.00	374	188
Nb	K12	0.00	2	28
Rh	K12	0.00	34	46
Rh	L1	0.00		275
Pd	K12	0.00	44	70
Pd	L1	0.00	3	262
Ag	K12	0.00	132	130
Ag	L1	0.00	1	267
Cd	K12	0.00	149	189
Cd	L1	0.00		252
Sn	K12	0.00	5828	383
Sn	L1	0.00	6	217
Sb	K12	0.00	490	469
Sb	L1	0.00	67	215
Pb	L1	0.00	86008	557
Pb	M1	0.00	525	304

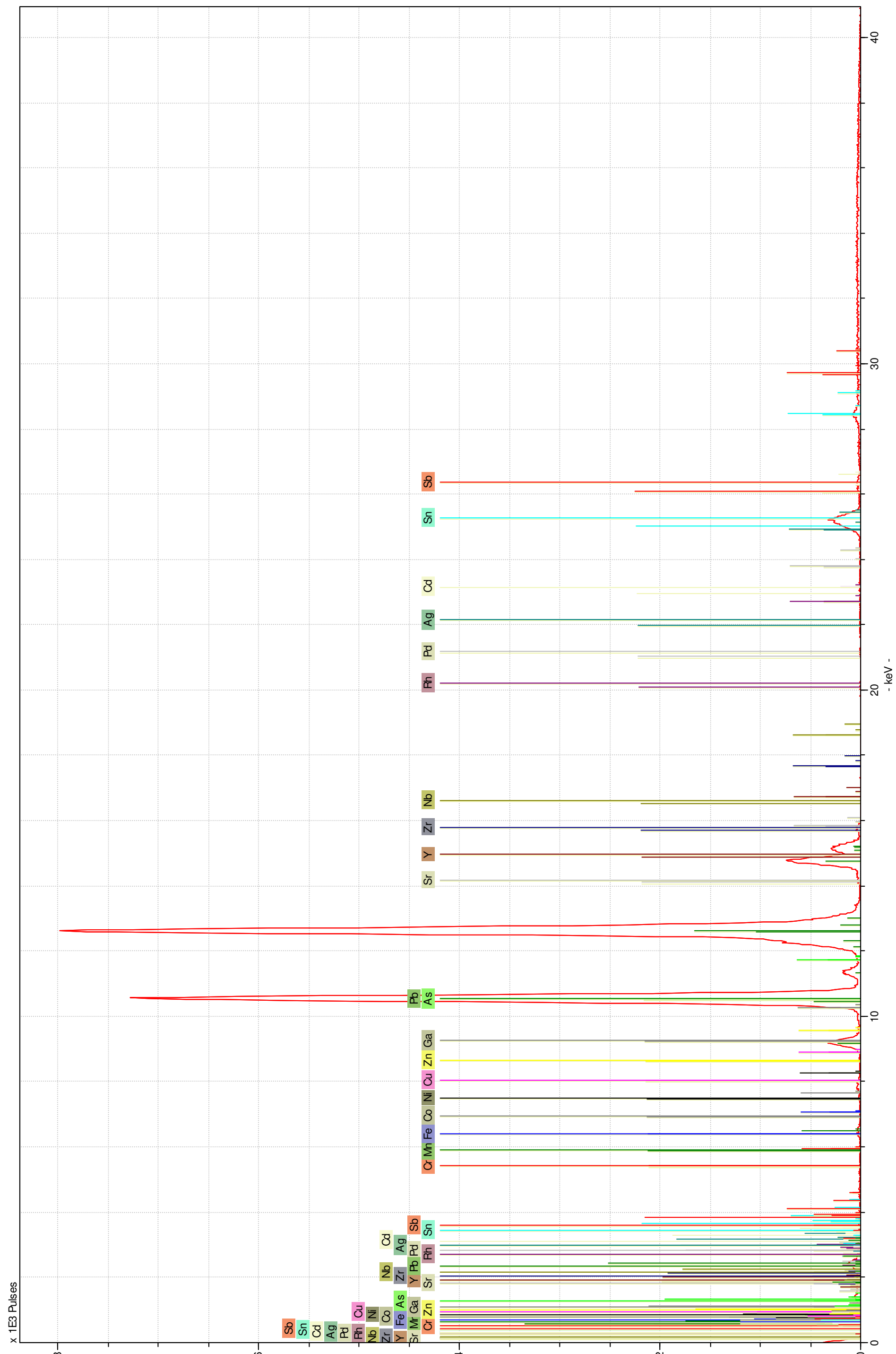

Figure 9. Spectra of Sample 46Gb13-9.

ARTAX - ELEMENT ANALYSIS

Listed at 3/12/2018 12:58:50 PM

Serial number:
Spectrum: 46gb13-10@120318_092540
Method: Lead balls (Bayes)
Count rate: 1418 cps
Voltage: 48 kV
Anode:
Optic:

Project: ArbucklesFort46Gb13.rtx
Meas.date: 3/8/2018 2:01:25 PM
Live time: 168 s
Dead time: 0.1 %
Current: 29 µA
Filter: Ti/Al/Cu
Atmosphere: Air

Element	Line	Sigma/	Net area	Backgr.
Cr	K12	0.00	68	83
Mn	K12	0.00	32	87
Fe	K12	0.00	230	93
Co	K12	0.00	38	103
Ni	K12	0.00	118	110
Cu	K12	0.00	192	134
Zn	K12	0.00	21	162
Ga	K12	0.00	701	246
As	K12	0.00	2339	564
Sr	K12	0.00	239	208
Y	K12	0.00	1663	263
Zr	K12	0.00	280	142
Nb	K12	0.00	3	18
Rh	K12	0.00	44	35
Rh	L1	0.00	1	249
Pd	K12	0.00	58	45
Pd	L1	0.00	4	232
Ag	K12	0.00	126	78
Ag	L1	0.00	3	230
Cd	K12	0.00	137	112
Cd	L1	0.00		211
Sn	K12	0.00	230	264
Sn	L1	0.00	8	168
Sb	K12	0.00	31	401
Sb	L1	0.00	21	167
Pb	L1	0.00	77671	515
Pb	M1	0.00	614	287

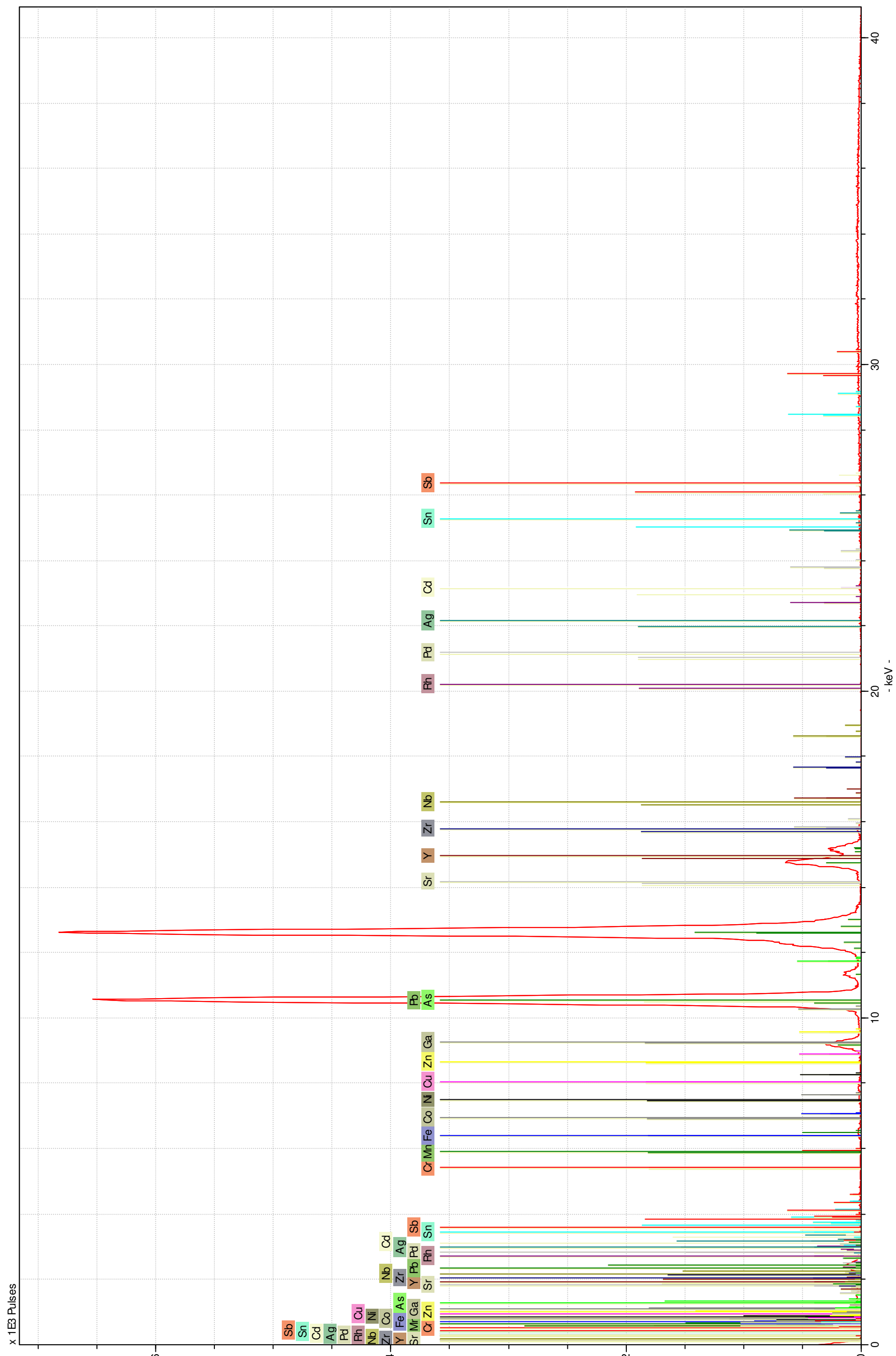

Figure 10. Spectra of Sample 46Gb13-10.

Sample	Ag K12	Cd K12	Cu K12	Fe K12	Ni K12	Pb L1	Pb M1	Pd K12	Rh K12	Sb K12	Sn K12	Zn K12	Diameter (mm)	Diameter (in)	Wt. (g)
46Gb13-1	71	144	277	431	85	91038	519	70	28	118	1554	53			12.6
46Gb13-2	97	69	155	308	82	36246	85	30	26	72	176	40			2.6
46Gb13-3	91	144	337	292	93	86168	563	70	35	487	5627	49	12.9	0.508	12.47
46Gb13-4	98	191	327	406	150	98320	590	93	33	50	1019	31			10.01
46Gb13-5	64	149	284	543	122	83680	403	72	49	7803	12156	22			9.74
46Gb13-6	50	127	275	256	85	75820	507	56	48	148	2454	21	8.92	0.351	3.8
46Gb13-7	102	245	370	355	124	107278	687	16	54	30	231	41			9
46Gb13-8	82	195	319	316	75	94528	632	61	26	33	209	47	12.12	0.477	10.2
46Gb13-9	132	149	284	354	114	86008	525	44	34	490	5828	48	10.39	0.409	6.6
46Gb13-10	126	137	192	230	118	77671	614	58	44	31	230	21			3.2

Sb/Sn	Cu/Pb	Sb/Rh	Sn/Rh	Ag/Rh	Cd/Rh	Cd/Sn	Pd/Rh	Sn/Sb	Ni/Rh	Cu/Rh	Zn/Rh	Sn/Pb
0.07593308	0.00304	4.2143	55.5	2.5357	5.1428571	0.092664	2.5	13.1695	3.03571	9.89286	1.89285714	0.017069795
0.40909091	0.00428	2.7692	6.7692	3.7308	2.6538462	0.392045	1.153846	2.44444	3.15385	5.96154	1.53846154	0.004855708
0.08654701	0.00391	13.914	160.77	2.6	4.1142857	0.025591	2	11.5544	2.65714	9.62857	1.4	0.065302665
0.04906771	0.00333	1.5152	30.879	2.9697	5.7878788	0.187439	2.818182	20.38	4.54545	9.90909	0.93939394	0.010364117
0.64190523	0.00339	159.24	248.08	1.3061	3.0408163	0.012257	1.469388	1.55786	2.4898	5.79592	0.44897959	0.145267686
0.0603097	0.00363	3.0833	51.125	1.0417	2.6458333	0.051752	1.166667	16.5811	1.77083	5.72917	0.4375	0.03236613
0.12987013	0.00345	0.5556	4.2778	1.8889	4.537037	1.060606	0.296296	7.7	2.2963	6.85185	0.75925926	0.002153284
0.15789474	0.00337	1.2692	8.0385	3.1538	7.5	0.933014	2.346154	6.33333	2.88462	12.2692	1.80769231	0.002210985
0.08407687	0.0033	14.412	171.41	3.8824	4.3823529	0.025566	1.294118	11.8939	3.35294	8.35294	1.41176471	0.067761138
0.13478261	0.00247	0.7045	5.2273	2.8636	3.1136364	0.595652	1.318182	7.41935	2.68182	4.36364	0.47727273	0.002961208

Scatterplot of Sb/Rh, Sn/Rh and Ag/Rh Ratios, Arbuckle's Fort Lead Ball Samples